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MASTER

TITLE: A NON-DESTRUCTIVE FIELD MEASUREMENT OF THE RATIO $^{235}U/^{238}U$ IN DEPLETED TO MODERATELY ENRICHED URANIUM

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A NON-DESTRUCTIVE FIELD MEASUREMENT OF THE RATIO $^{235}U/^{238}U$ IN DEPLETED TO MODERATELY ENRICHED URANIUM

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ABSTRACT

The exploration of a natural reactor site is expedited by prompt measurement of ^{235}U to ^{238}U ratios near the mining operation. An instrument has been constructed which uses the relative fission rates of ^{235}U and ^{238}U in fast and moderated neutron spectra to measure the isotopic ratio. This device can be placed in the field and allows continuous monitoring of ore as a rich deposit of uranium is mined. With rapid return of isotopic information to the operator it is possible to locate a fossil reactor before it has been destroyed.

The relative fast neutron and slow neutron fission rates induced in uranium which is depleted to moderately enriched in ^{235}U may be used to measure the isotopic ratio $^{235}U/^{238}U$ quickly and non-destructively with a relative error of a few percent. When a neutron source such as ^{252}Cf is used, the measurements may be made in the field.

The discovery of the OKLO natural fission reactor has stimulated interest in a search for other natural reactors. If evidence for a reactor were to be found in the course of the actual mining of a rich deposit, definitions of the regions of greatest interest and optimization of a sample-collection strategy would be expedited by a fast field technique for measuring the isotopic ratio $^{235}U/^{238}U$ in ore samples. An instrument for providing such data at OKLO was developed and used in the field by French scientists.[1] It is based on measurement of the 186 KeV gamma ray in the ^{235}U decay spectrum and a transmission measurement to obtain the absolute concentration of uranium.

An instrumental technique for quickly identifying depleted samples ($^{235}U/^{238}U<0.6\%$) without requiring dissolution and purification of uranium has been developed at Los Alamos. It is based on the ratio of fissions induced in an ore sample by unmoderated and moderated neutrons from a ^{252}Cf source. With the existing neutron sources (three sources of 300 μg each) and a high counting efficiency for fission fragments (∿100%) the analysis requires about forty minutes using an ore of greater than 20% U_3O_8 content. The standard deviation of the isotopic ratio measured on 23 samples of natural ore from Canada and Australia is 4% relative. Although the precision of the technique is not high it is sufficient to identify the main reactor zone regions in an OKLO type deposit without the need for a supporting chemistry laboratory. In addition, the features of the instrument which recommend it for field use are:

1. It can be carried on a light truck
2. The neutron source is reliable and reproducible
3. Supporting materials can be obtained at the site
4. Sample preparation is minimal
5. The results are available on a sufficiently rapid time scale so that a minimum of interference with an ongoing mining operation should be necessary.
6. The procedure can be carried out with field personnel.

Estimates were made to determine if a measurement based on fission counting of the ore using moderated and unmoderated neutrons would give reasonable isotopic sensitivity. Monte Carlo calculations [Figure 1] of normal U_3O_8 response to moderated and unmoderated neutrons indicated that in the proposed cylindrical geometry [Figure 2], with the uranium at the center of the cylinder 98% of the fissions in the unmoderated spectrum would be in the ^{238}U and that 97% of the fissions would be in ^{235}U in the moderated spectrum. Thus the ratio of fission counts from the two neutron spectra are essentially proportional to the ^{235}U abundance.

A schematic diagram of the instrument as built is illustrated in Figure 3. In use the neutron sources are removed from the shipping cask remotely and placed in the source tube in the water shield. After an ore sample is loaded into the counter, the ^{252}Cf sources are lowered around the sample. Measurements are made with the counter dry and then filled with water. A complete measurement can be made in 40 minutes on an ore of U_3O_8 content of 20% or better to a precision of 0.3% in the statistics of an individual count.

Calibration was accomplished with standards made by mixing NBS (National Bureau of Standards) U_3O_8 of natural isotopic composition and U_3O_8 depleted in ^{235}U (5700 ^{238}U: 1 ^{235}U).

A least squares fit to the data of Figure 4 which was obtained using the above standards gives an equation for the line of $y = 1.2619 + 616.4\ x$, where y is the ratio of moderated count divided by unmoderated count. The value for x is the weight ratio of $^{235}U/^{238}U$. The precision of the data gives a standard deviation of ±4% for each point. The calibration curve over the range investigated is adequately described by a linear fit.

Three samples from the OKLO reactor were then examined. Results are shown in Figure 4 and in Table I with a comparison to the isotopic composition as determined by mass spectrometry. Further measurements using this instrument have been done on ore samples obtained from the Maurice Nabarlek deposit in northern Australia and the deposits at Key Lake and Marnice Bay in Saskatewan, Canada. Twenty-three samples have been measured from these deposits. The results are shown in Table II. No ratios have been found outside the ±2σ range.

REFERENCES

[1] Boyer, R. L., Guery, M.P., Renson, C.

Appareil De Measure De Teneur Isotopique De L'Uranium, Utilise Sur Le Site D'Oklo, Le Phenomene D'Oklo, Compte rendu d'un Colloque Libreville 27 Juin 1975 IAEASM204/22

TABLE I

OKLO ISOTOPE RATIOS

$^{235}U/^{238}U$

Source	Present Measurement		Mass Spectrometry
OKLO			
401/3	4.18×10^{-3}	$\pm.13 \times 10^{-3}$	4.05×10^{-3}
404/4	5.67×10^{-3}	$\pm.14 \times 10^{-3}$	5.55×10^{-3}
433/3	4.42×10^{-3}	$\pm.19 \times 10^{-3}$	4.52×10^{-3}

TABLE II

AUSTRALIAN AND CANADIAN

SAMPLE RATIOS

$^{235}U/^{238}U$ Ratio

Source

NABARLEK	KEY LAKE	MAURICE BAY
7.02×10^{-3}	7.18×10^{-3}	7.08×10^{-3}
6.90×10^{-3}	6.95×10^{-3}	7.01×10^{-3}
7.36×10^{-3}	7.32×10^{-3}	
7.17×10^{-3}	7.25×10^{-3}	
7.28×10^{-3}	6.62×10^{-3}	
7.79×10^{-3}	6.68×10^{-3}	
6.93×10^{-3}	7.05×10^{-3}	
7.20×10^{-3}		
7.38×10^{-3}		
7.61×10^{-3}		
7.63×10^{-3}		
6.85×10^{-3}		
7.15×10^{-3}		
7.76×10^{-3}		

Mean of all Samples 7.18×10^{-3}

$\sigma \pm 0.31 \times 10^{-3}$

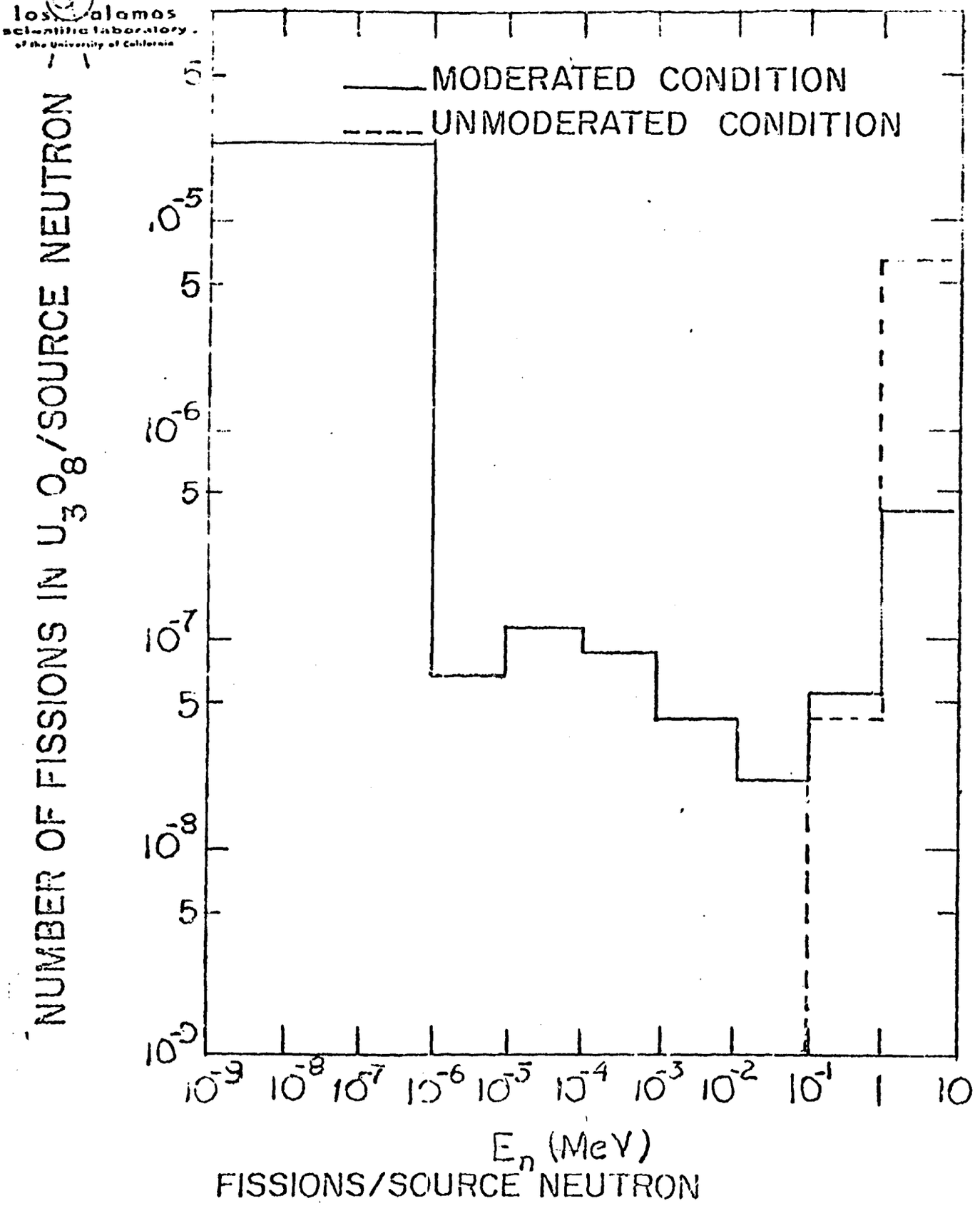
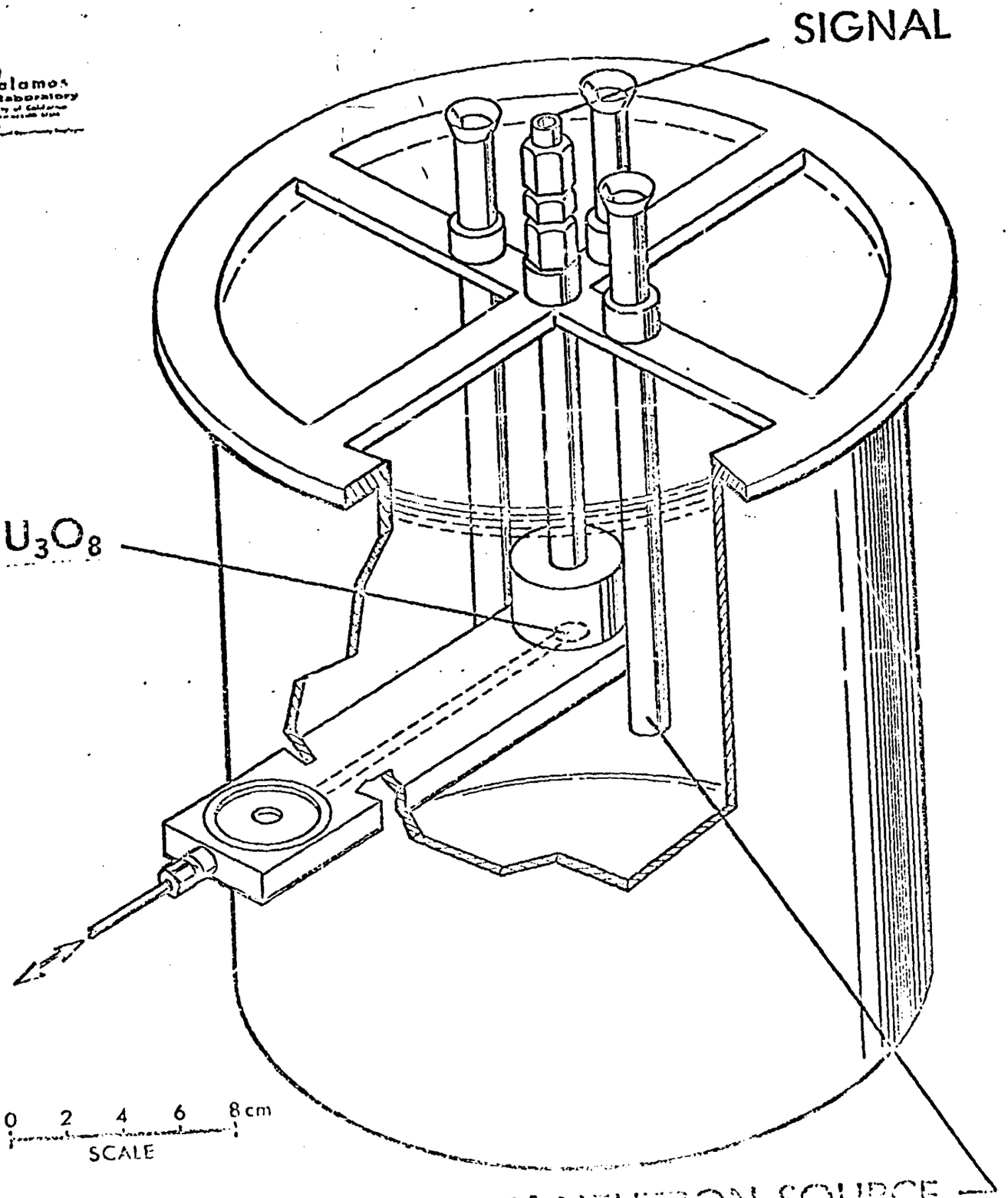
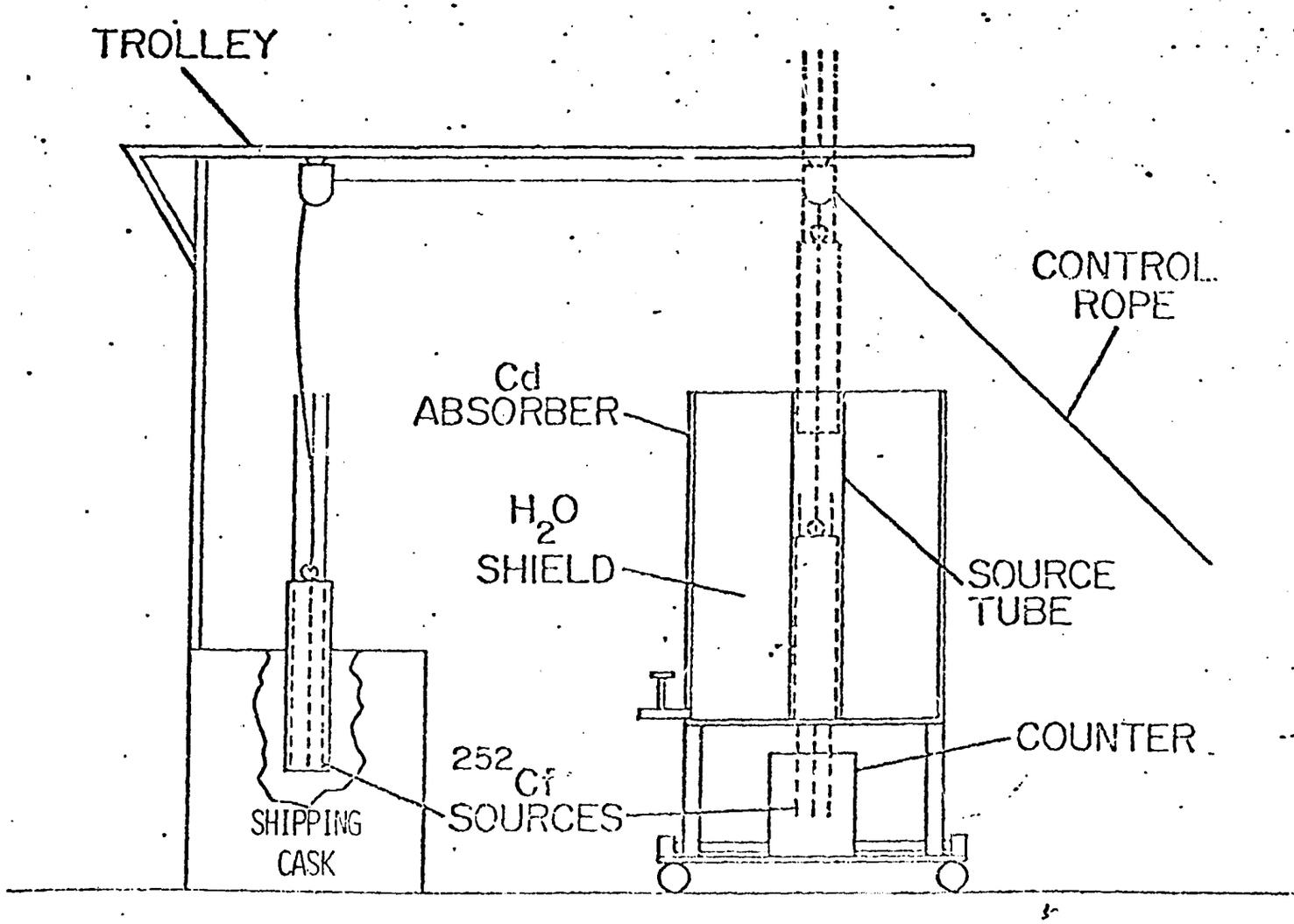

MODERATED CONDITION
UNMODERATED CONDITION
NUMBER OF FISSIONS IN U_3O_8/SOURCE NEUTRON
10^{-5}
5
10^{-6}
5
10^{-7}
5
10^{-8}
5
10^{-9}
10^{-9} 10^{-8} 10^{-7} 10^{-6} 10^{-5} 10^{-4} 10^{-3} 10^{-2} 10^{-1} 1 10
E_n (MeV)
FISSIONS/SOURCE NEUTRON

FIGURE I



COUNTER

FIGURE 2



CROSS SECTION OF ISOTOPE METER

FIGURE 3

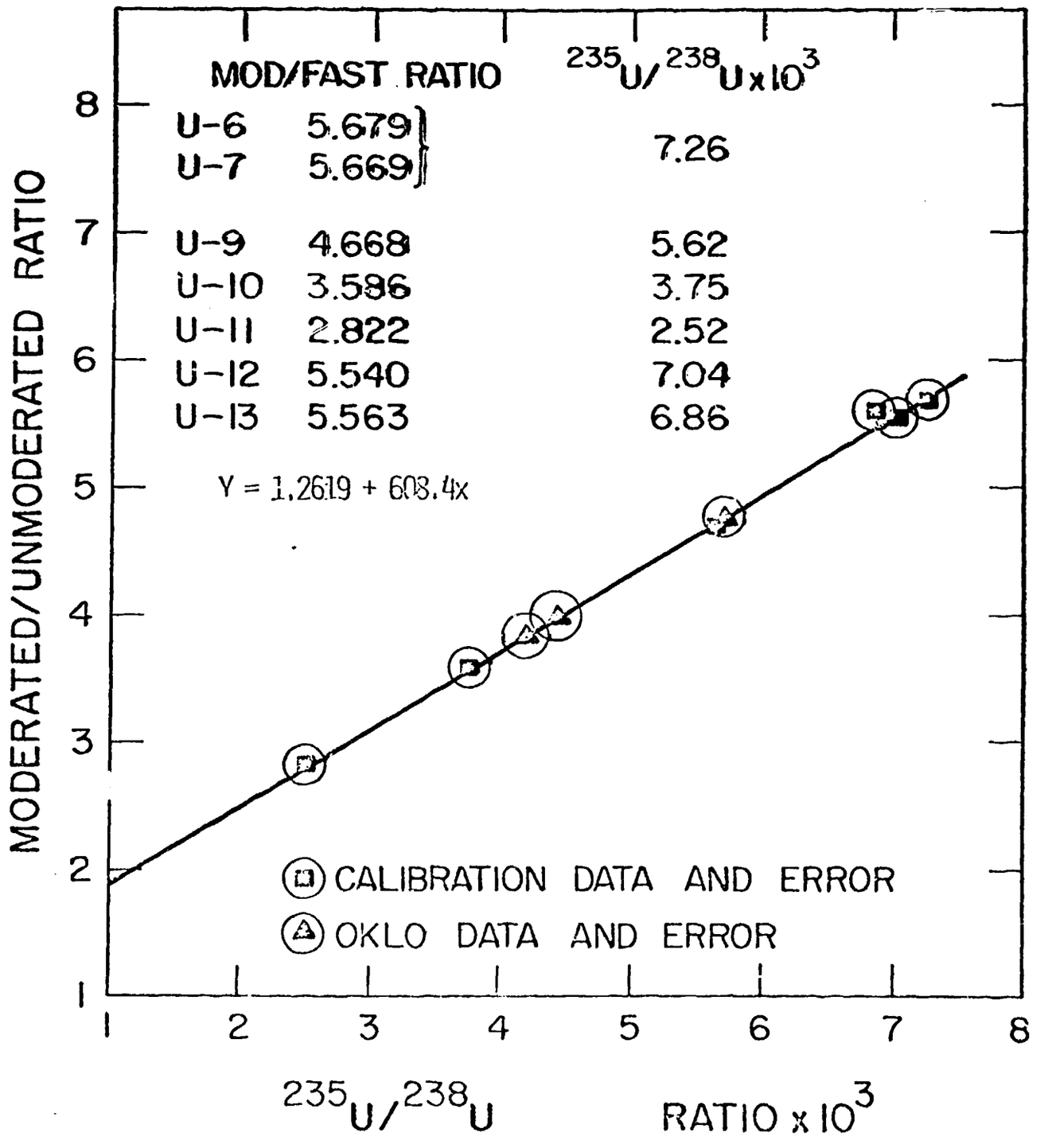

MOD/FAST RATIO
$^{235}U/^{238}U \times 10^3$
U-6 5.679
U-7 5.669
7.26
U-9 4.668 5.62
U-10 3.586 3.75
U-11 2.822 2.52
U-12 5.540 7.04
U-13 5.563 6.86
Y = 1.2619 + 608.4x
CALIBRATION DATA AND ERROR
OKLO DATA AND ERROR
MODERATED/UNMODERATED RATIO
$^{235}U/^{238}U$ RATIO x 10^3

FIGURE 4